Neutron Small Angle Scattering on Liquid Helium in the Temperature Range 1.5-4.2 K

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Abstract

The small angle neutron scattering (SANS) from liquid helium at saturated vapour pressure in the temperature range from 1.5 to 4.2 K was measured with the instrument D22 of the ILL Grenoble at a wavelength of 4.6 Å. The zero angle cross section is monotonically decreasing with decreasing temperature and does not show any singularity at the lambda-point. On the other hand, we observe a change of the slope of the temperature dependence of the second momentum of the pair correlation function at the lambda-point that reflects the transition of liquid to the superfluid state.

1 Introduction

Thermal neutrons are ideally suited to probing the excitations in condensed-matter systems, as their wavelengths $0.5 < \lambda < 10$ Å are comparable to the interatomic distances, and their energies are of the same magnitude as the excitations. In particular, the small-angle neutron scattering (SANS) gives valuable information on thermal fluctuations in liquids.

The only previous SANS experiment on liquid helium has been performed by Egelstaff and London long ago, in 1953 [1]. Unfortunately, there are only a few experimental points below the lambda point, and it is difficult to come to a definite conclusion about the small-angle scattering behaviour in a wide temperature region. Modern high-flux reactors and advanced instruments have increased the detection sensitivity by orders of magnitude. Additionally, previous detail experiments on the static structure factor of helium-4 have been performed only for the momentum transferred above $1\,\text{Å}^{-1}$ [2, 3].

This was a motivation for us to remeasure SANS from liquid helium in a wide temperature range.

2 Experiment

The experiments were carried out at the instrument D22 of the Institut Laue-Langevin (ILL), Grenoble, France. We used a standard ILL Orange Cryostat modified for SANS with quartz windows. We filled the inner part of the cryostat, which is delimited by transparent quartz windows at the neutron entrance and exit sides, with liquid helium by condensing it into the sample volume. We then lowered the helium temperature first by opening the cold valve, and when we came close to the phase transition to He-II, by directly pumping on the helium.

The experiments were performed with $4.6\,\text{Å}$ neutrons. The sample-to-detector distance was $2.8\,\text{m}$ (just avoiding shadowing on the detector that was centred with respect to the beam). The "collimation" distance, i.e. the distance between the end of the neutron guide of $40\,\text{mm}$ (wide) by $55\,\text{mm}$ (high) and the sample, was $5.6\,\text{m}$. The sample aperture had a diameter of $10\,\text{mm}$. The scattering from the empty cryostat at low temperature, and the neutron and electronic noise were measured in order to correct the data for backgrounds. The data were radially averaged and the container and noise contributions were subtracted by standard programs; they were put on an absolute scale by normalizing with the scattering from $1\,\text{mm}$ of water. We also checked by measurements with a detector setting of $14\,\text{m}$ that there was no central increase in the helium scattering pattern.

3 Results and Discussion

Figure 1 shows radially averaged scattered neutron intensities versus momentum transfer for some helium temperatures and water normalized scattering patterns with sample background subtracted.

The momentum transferred is calculated on the assumption that the neutron scattering is elastic and is related to the full scattering angle θ through the expression

$$|Q| = \frac{4\pi}{\lambda}\sin(\theta/2), \tag{1}$$

where λ is the wavelength of the neutron.

Although the background from the cryostat is rather small in comparison with the intensity scattered from helium, we see an increase of the background intensity at low momentum transfer ($Q < 0.03\,\text{Å}^{-1}$) and, as a consequence, a decrease of an intensuty of scattered from helium neutrons as seen in Fig.1. A reasonable explanation is that the small-Q scattering from the cryostat is a result of the existence of dust, cracks, scratches, that are small in size, on the surfaces of the quartz windows. Special experiments are needed to clarify this situation, and that is why we analyse only experimental data in the Q-range $(0.03 \div 0.3)\,\text{Å}^{-1}$.

The essential quantity in the description of a liquid is its structure factor $S(Q)$ that
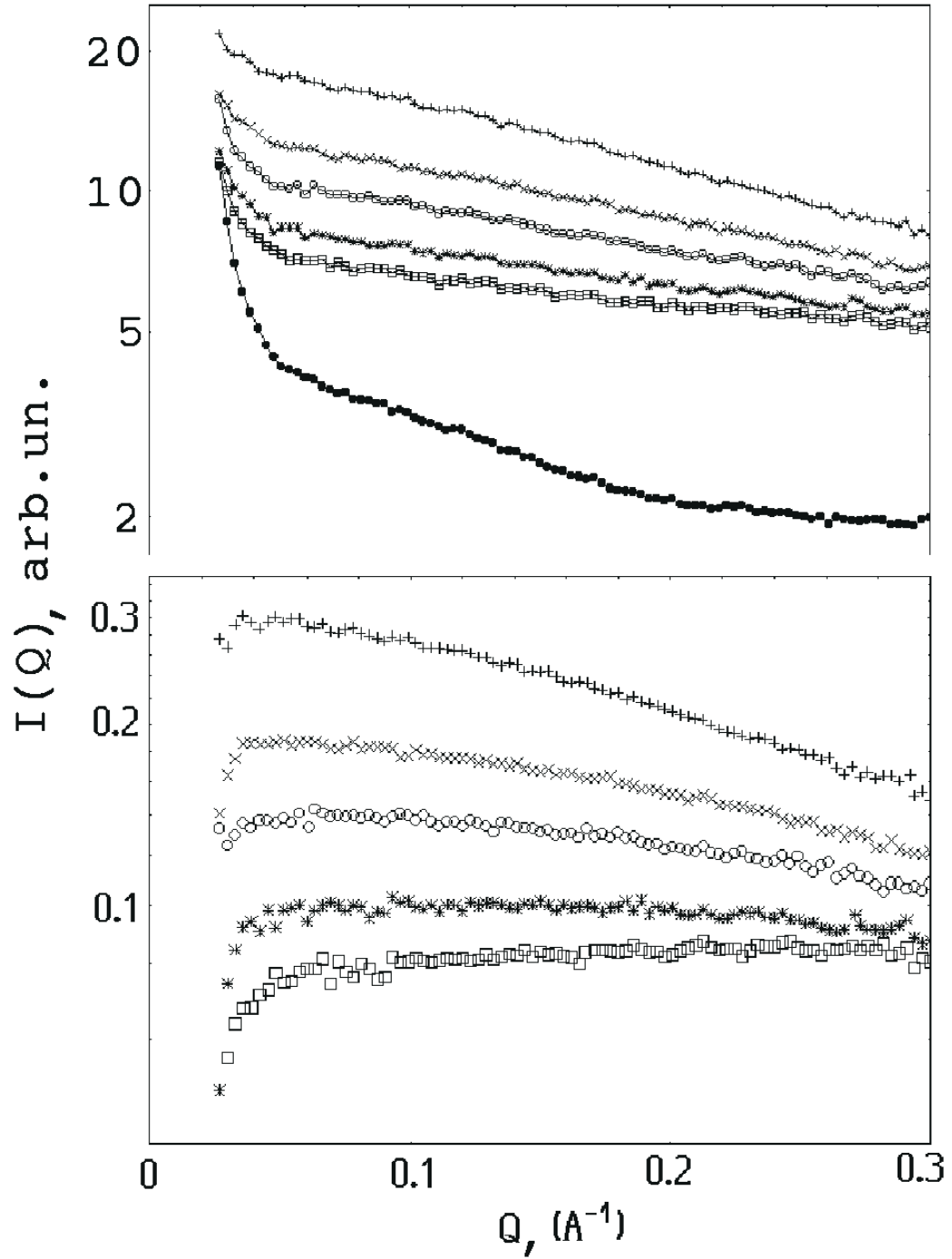
Figure 1: *Upper set of curves:* Radially averaged intensity of scattered neutrons (in arbitrary units) *versus* momentum transfer at some helium temperatures: + — 4.09 K, × — 3.54 K, ○ — 3.04 K, * — 2.18 K, □ — 1.54 K, full □ — empty cryostat. *Lower set of curves*: water normalized scattering patterns with sample background subtracted.

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is a measure of the correlation between positions of the atoms in fluid. The macroscopic cross section $d\Sigma/d\Omega$ of neutron scattering is related to the static structure factor $S(Q)$ by

$$\frac{d\Sigma}{d\Omega}(Q) = \frac{\sigma_b}{4\pi}\rho_{\text{at}}S(Q), \tag{2}$$

where σ_b is the bound atom scattering cross section of helium (= 1.172 barn) and ρ_{at} is the atomic density of helium.

According to Goldstein [4], at finite temperature the structure factor is given by

$$S(Q) = n_0 k_{\text{B}}T\chi_T + \sum_{n=1}^{\infty}(-1)^n Q^{2n}r_g^{2n}[(2n+1)!]^{-1}, \tag{3}$$

where n_0 is the number density, k_{B} the Boltzmann constant, χ_T the isothermal compressibility, and r_g^{2n} is the moment of the pair correlation function $g(r)$ defined by

$$r_g^{2n} = n_0 \int r^{2n}[1 - g(r)]d^3r. \tag{4}$$

In particular, to order Q^2 Eq.(3) is just

$$S(Q) \simeq n_0 k_B T\chi_T - r_g^2\frac{Q^2}{6}, \quad S(0) = n_0 k_B T\chi_T. \tag{5}$$

This approximation looks like the Guinier approximation of the decay of the intensity near $Q = 0$

$$I(Q) = I(0)\exp(-Q^2 R_g^2/3) \simeq I(0)(1 - Q^2 R_g^2/3), \tag{6}$$

where R_g is the radius of gyration.

On the other hand, as it shown by Ornstein and Zernike, in the range of critical point the intensity of scatterred neutrons is described as

$$S(Q) = \frac{S(0)}{1 + r_c^2 Q^2} \simeq S(0)(1 - r_c^2 Q^2). \tag{7}$$

We see that there is a direct relation between r_c, R_g and r_g. The critical temperature of helium equals $5.2\,\text{K}$, and thus we can definitely say that at temperature around $4\,\text{K}$ the value of R_g, as well as r_g, reflects correlation between helium atoms and we can consider them as correlation radius. As to smaller temperatures, the physical meaning of these quantities not so apparent.

The parabolic behaviour of the intensity of scattered neutrons at small Q is clearly seen in the experiment (Fig.2).

It is necessary here to note that we have not take into account the scattering from zero-point fluctuations that leads to the linear function of Q that has been first obtained by Feynman [5], but this process takes place at lower temperatures.
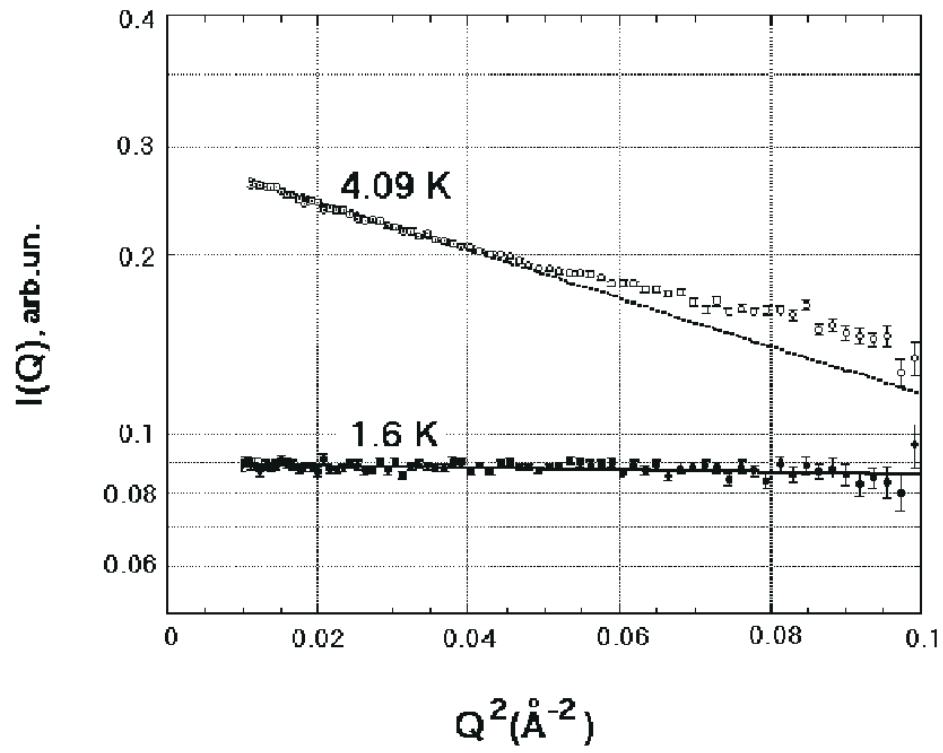
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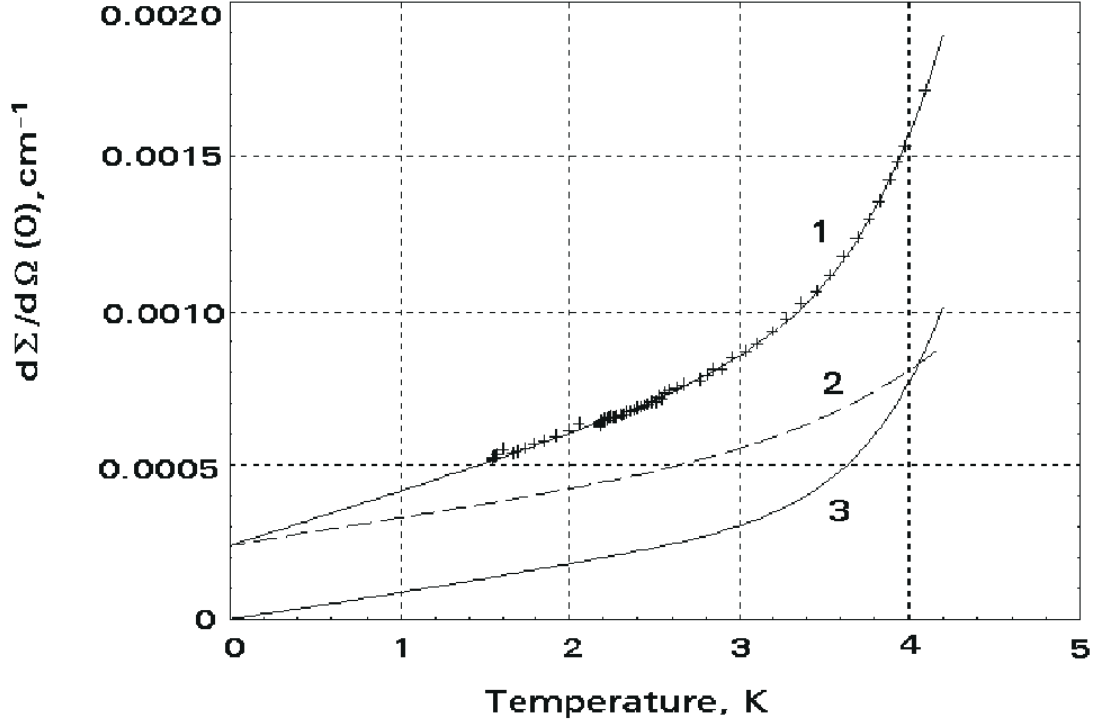Figure 2: Intensity of scattered neutrons versus Q^2.

Figure 3: 1 — Zero-angle cross sections (+), solid line — its fit with linear Q-dependence at low temperatures; 2 — fit of measured background; 3 — theoretical zero-angle cross section.

We calculated the zero-angle cross section by extrapolation of the scattered intensity to Q=0, and the results are shown in Fig.3. It is seen that the zero angle cross section is monotonically decreasing with decreasing temperature and does not show any peculiarity at the lambda-point. In the same figure we show the theoretical zero angle cross section $S(0)$ calculated using the known data for the temperature dependence of the helium density and the isothermal compressibility [8]. At temperatures below the lambda-point both density and isothermal compressibility are practically independent of the temperature, and thus the zero-angle cross section is proportional to the temperature that allows it to fit linearly at low temperatures. As seen, there is a temperature dependent background, but we don't know its origin.

We have measured SANS with a temperature interval 0.1-0.2 K that has provided the first comprehensive data on the correlation radius in the normal and superfluid state of helium. The results are shown in Fig. 4. The presented in Fig.1 data were corrected for constant background that corresponds to $d\Sigma/d\Omega(0)$ at zero temperature. It is seen that the temperature dependence of the correlation radius behaves like for a normal liquid, i.e. it
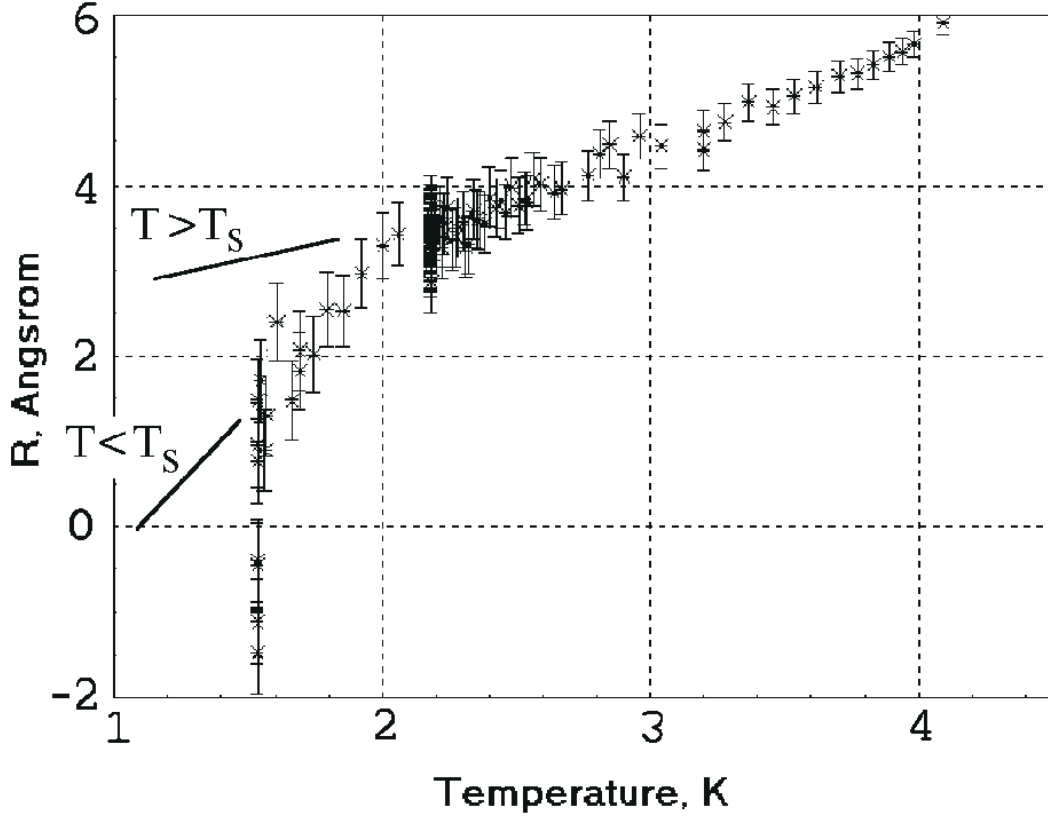
Figure 4: Radius of gyration R as a function of temperature. The negative sign of the radius means the change of the derivative of the function $I(Q^2)$. For clarity the slope of $R(T)$ in the vicinity of the temperature of the superfluid transition T_s is shown by solid lines.

decreases with decreasing temperature. However, at the superfluid transition temperature (2.2 K) we see a pronounced change of its slope.

At a qualitative level the observed loss of spatial correlations in liquid helium is due to a rearrangement of atoms to create the necessary spaces for delocalisation to occur. Nevertheless, this intuitive explanation has not theoretical basis. For instance, using Monte Carlo method Mayers [6] argued that the link between spatial correlations and Bose-Einstein condensate fraction is a geometrical consequence of the hard-core repulsion between atoms. The excitation of rotons has also been proposed by Masserini *et al.* [7] as an alternative explanation for the increase in spatial correlation as the temperature is rased in the superfluid state.

In this connection, we have to emphasize that in our SANS experiment the distance scale probed is greater than interatomic distances, and the correlation length obtained characterizes long-range order in liquid helium whereas in all previous experimental and theoretical works the behaviour of the first peak in the $S(Q)$ was considered, i.e. short-

range correlations at Q-value about 2 angstrom^{-1}. In our SANS experiment we deal with $Q < 0.3\,\text{Å}^{-1}$. As opposed to previous experiments we have studied long-range correlation between helium atoms.

4   Acknoledgements

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